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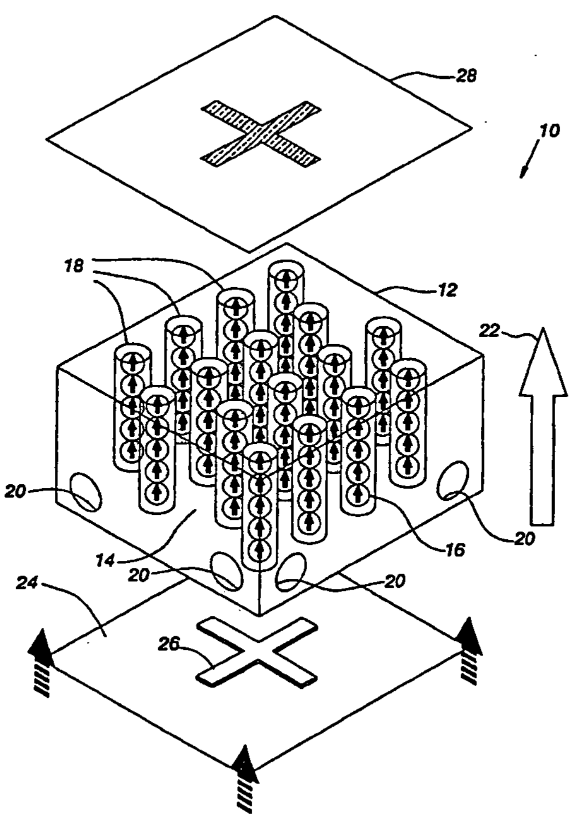
(54) Title: ACOUSTIC FLUID JET METHOD AND SYSTEM FOR EJECTING DIPOLAR GRAINS



(57) Abstract: A method and apparatus (10) for ejection of nanometer length-scale, dipolar grains (16) from a host liquid (14), such as a dilute ferrofluid with grains suspended in water (e.g., γ-Fe_2O_3). In a preferred embodiment, the host liquid is subjected to a strong, homogeneous magnetic field (22) directed perpendicular to an exit surface of the liquid, thereby causing columns of dipolar grains (16) extending parallel to the field direction to form. An acoustic impulse is initiated at the base of the container (12) and propogates as a non-dispersive solitary wave pulse with sufficient energy to overcome surface tension and eject a ferrofluid grain nearest to the exit surface of the liquid. In another preferred embodiment, a rapid series of acoustic impulses constructively interfere with each other to eject a plurality of grains nearest the exit surface. The method and apparatus of the invention are useful in the design of a nozzle-free ink-jet printer (10) of unparalleled resolution.

WO 01/17781 A1

ACOUSTIC FLUIT JET METHOD AND SYSTEM FOR EJECTING DIPOLAR GRAINS

Technical Field

This invention relates generally to methods of rapid ejection of nanometer length-scale, dipolar grains from a host liquid. The ejection procedure can only be effected in the presence of an electromagnetic field, which can self-assemble the dipolar grains inside a host liquid in chains or columns, and by imparting a suitable impulse at the base of the chains or columns to eject a grain at a time from the surface of the chain. The application of this concept is in high-speed, high-resolution printing; more specifically, it relates to inkjet printing, and even more particularly, to a nozzle-free, nanometer length-scale resolution method of inkjet printing.

Background Art

In general, ink jet printers function by projecting charged droplets of ink from a nozzle and through an electrostatic field. By controlling the charge imparted to each droplet, one can control the placement of the droplets on the target media (e.g., paper). The ink droplets may be applied continuously or in a demand pulse manner. Precise control of the droplets from the time of creation to the time of deposit on paper or other media is essential. The quality of a printed image, measured as resolution, is a function of the number of ink drops that can be placed per unit area (usually expressed as dpi, or dots per inch). A higher resolution results in sharper, more detailed images. Various printhead structures and drive circuits have been developed to improve control of the printing and to improve resolution. The resolution of ink jet printers is generally 300 dpi. Since resolution is directly related to drop size (i.e., the smaller the diameter of the drop, the higher the possible resolution), regardless of the degree of control achieved, resolution has heretofore always been limited by the diameter of the nozzle in ink jet printing.

Examples of inkjet printing improvements in method and apparatus are well documented in the patent art. In 1974, for example, Richard M. Hecht and Hubert D. Faulkner received United States Patent No. 3,852,772 (Dec. 3, 1974) for an invention

entitled, "Mechanically Cycled Ink Jet Printer". This invention used a mechanically cycled ink jet printer nozzle, through which a stream of ink passes and from which there issues a stream of droplets of inks. Lateral vibration caused the stream to follow a cyclic trajectory, and selective charging caused some of the droplets to impinge on the target media, while other uncharged droplets were deflected to a catcher.

United States Patent No. 4,387,383 (Sayko) (June 7, 1983) discloses an ink jet print head having a plurality of nozzles. Piezoelectric elements (one for each nozzle) in direct contact with the fluid are pulsed on demand and in succession to cause rapid volume changes in the fluid chamber to initiate pressure waves and eject droplets of ink. This patent references many developments in the field of ink jet print heads. Quoting from the patent at columns 1 and 2:

> "Representative prior art in the field of ink jet print heads includes U.S. Pat. No. 3,373,437, issued to R.G. Sweet et al. on Mar. 12, 1968, which discloses a fluid droplet recorder with a plurality of jets and wherein a common fluid system supplies ink to an array of side-by-side nozzles.
>
> U.S. Patent No. 3,683,212, issued to S.I. Zoltan on August 8, 1972, discloses an electro-acoustic transducer coupled to liquid in a conduit which terminates in a small orifice through which droplets of ink are ejected.
>
> U.S. Patent No. 3,750,564, issued to H. Bettin on August 7, 1973, discloses a multiple nozzle ink jet print head having an ink chamber with opposed electrodes and insulating partitions to define capillary chambers. Ink drops are initiated by electrical forces of attraction and repulsion between the charged writing fluid in a capillary channel and electrodes of opposite polarity mounted on either end of the capillary channel.
>
> U.S. Patent No. 3,832,579, issued to J.P. Arndt on August 27, 1974 discloses a pulsed droplet ejecting system wherein an electro-acoustic transducer applies a pressure pulse to the liquid in

a reflection-free section of the transducer and sends a pressure wave to the nozzle to cause ejection of an ink droplet.

U.S. Patent No. 4,005,440, issued to J.R. Amberntsson et al. on January 25, 1977, discloses a printing head of smaller size and wherein the openings of the capillary tubes are located closer to one another.

U.S. Patent No. 4,032,928, issued to J.T. White et al. on June 28, 1977, discloses a wide band ink jet modulation having a base and a nozzle plate spaced therefrom with a transducer, an electrode and a diaphragm axially positioned to cause droplets of ink to be ejected from an ink chamber and through the nozzle in the plate.

U.S. Patent No. 4,096,626, issued to C.E. Olsen et al. on June 27, 1978, discloses a method of making a multi-layer laminated charge plate for an ink jet printer wherein etched layers of photosensitive glass are provided with slots in the thickness of the layers for conductors.

U.S. Patent No. 4,128,345, issued to J.F. Brady on December 5, 1978, discloses a fluid impulse matrix printer having a two-dimensional array of tubes in a 5x7 matrix to print a complete character at a time.

U.S. Patent No. 4,158,847, issued to J. Heinzl et al. on June 19, 1979 discloses a piezoelectric operated print head having twin columns of six nozzles.

U.S. Patent No. 4,189,734, issued to E.L. Kyser et al. on February 19, 1980 discloses a writing fluid source feeding drop projection which ejects a series of droplets through a column of nozzles with sufficient velocity to traverse a substantially straight trajectory to the record medium."

United States Patent No. 4,947,184 (Moynihan) (August 7, 1990) relates to a method and apparatus for eliminating nucleation sites in a pressure chamber of an ink jet system. In particular, the patent teaches coating the pressure chamber with a smooth, conforming layer of a coating material, such as a xylylene polymer material, wettable by the ink to eliminate nucleation sites in the surfaces forming the walls of the chamber and inhibiting formation of bubbles from dissolved air contained in ink within the chamber.

United States Patent No. 5,087,930 (Roy et al.)(February 11, 1992) discloses an extremely compact ink jet print head having an array of closely spaced nozzles supplied from densely packed ink pressure chambers by way of offset channels. The system is intended for use in drop-on-demand ink jet printing systems.

A creative method of increasing resolution in an ink jet printing system is disclosed in United States Patent No. 5,677,714 (Klassen et al.) (October 14, 1997). The purpose of the invention is to reproduce an image at a 600 dpi resolution using an ink jet printer having only a 300 dpi resolution capability. To achieve this purpose, the method of the invention maintains the edges of the pixel image and uses a checkerboard pattern for all interior pixels other than the second pixel and the second last pixel of each pixel row. The method is neighbor insensitive by using a checkerboard pattern or mask to determine the state of the interior pixels. After turning "off" the respective pixels, ink drops are fired from the ink jet printer at areas corresponding to the remaining "on" pixels, thereby achieving a higher resolution.

Thus, it is seen that the art is full of attempts to improve the quality, resolution and speed of ink jet printing systems. It appears that all of these attempts, however, start with the premise that an inkjet printer must include one or more nozzles. Nozzles are used for passing the ink at high speeds and for breaking up the inkjet into tiny droplets. Smaller the nozzle diameter, larger is the velocity needed to pass the ink through the same and hence improved are the prospects of producing proportionately small droplets. None of the patents cited above even remotely suggests that it might be possible to greatly improve resolution by eliminating the nozzle altogether. Thus, it is necessary to explore the development of inkjet printers that do not use nozzles, and hence are capable of attaining unprecedented size resolution.

There is no known method of ejecting nanometer sized grains out of a colloidal liquid, where such particles can exist in stable form, i.e., without being precipitated. Typically, these grains carry a sticky skin of the host liquid. The host liquid can be water-based or oil-based and can, in some cases, be colored. The ejecting grain can hence be considered as a "colored nanodrop." Such a grain can be embedded on to a plurality of surfaces such as office-quality paper, newspapers, paper used in making currency, surfaces that are appropriate for preparing ultra-dense microfiche, metal and semiconductor surfaces such as in casings of computer chips and others.

Few modern technologies are capable of making nanometer sized drops with regard to printing applications. One of the established techniques is given below.

K. Kim and C.K. Ryu, U.S. Patent No. 5,344,676 (Sept 6, 1994) have introduced a method and an apparatus for producing nanodrops and nanoparticles and film deposits therefrom. These inventors have used an electrostatic spraying technique that is capable of producing liquid drops with diameter less than 1 micron (being called, "nanodrops"). See for example in K. Kim et al., "Generation of charged drops of insulating liquids by electrostatic spraying," J. Applied Physics Vol. 47, No. 5, (May 1976) pp 1964-1969 (1976) and J. Woosley et al., "Electrostatic spraying of insulating liquids: H_2," IEEE Trans. Ind. Appl. Vol. 1A-18, No. 3 (May/Jun 1982) pp. 314-320. In this approach, a liquid precursor is placed in an open-ended tube within which is a solid electrically conductive needle, which protrudes beyond the open end of the tube. Surface tension of the liquid at the tube end prevents the liquid from flowing from the tube. Mutually repulsive electric charges are injected into the liquid through the needle, causing the surface tension to be overcome to produce a plurality of liquid jets, which break into nanodrops. This method is capable of making nanodrops but is not suitable for controlled ejection of grains of given sizes and for producing drops that are appropriate for printing and nanoscale writing related applications.

Disclosure of Invention

Application of controlled ejection of nanodrops in printing technology is addressed below.

The proposed printer exploits dipolar colloids (such as water based γ-Fe_2O_3 ferrofluids) that can be colored and used as ink. The grains in the colloid can be of any suitable size (between 50 and several hundred angstroms) and should preferably be approximately monodisperse. They also must be highly responsive to applied magnetic or electric fields and align in chains or columns in a direction parallel to that of the applied field. Typical ferrofluid grains in the above-mentioned system possess a magnetic moment of about 20,000 Bohr magnetons. To accomplish chain formation, the ink container must be sufficiently shallow in depth and the concentration of the colloidal grains must be small (few percent) and a homogeneous electric (for electrorheological colloids) or magnetic (for magnetorheological colloids) field must be established. Chain formation occurs in a direction that is parallel to that of the applied field. A suitable way to accomplish chain formation is to place the colloidal fluid in a container with a commercially available material with many vertical pores, each with diameters of some 10-20 nanometers. In such a system, each chain may form in each vertical pore and allow the ejection of grains from each pore upon the initiation of an impulse. The system will require a several microseconds to recover after each impulse or sequence of rapid impulses, before chains naturally reassemble.

Upon imparting a nonlinear acoustic impulse across a very short time (few tenths of a microsecond) at the bottom of the container using strikers with embossed designs of the imprint to be made on some surface, it is possible to generate solitary wave-like impulses through the chains of colloidal grains. The initial impulse can be arranged such that when the solitary wave-like impulse reaches the surface of the liquid it possesses sufficient energy to overcome surface tension of the liquid and eject the surface grain from the liquid. The ejected grain can be directed toward a piece of properly coated printer or other appropriate quality paper, a material surface or some other object and an imprint can be made.

Brief Description of Drawings

Fig. 1 is a schematic diagram of a ferrofluid printer formed in accordance with a preferred embodiment of the present invention.

Fig. 2a is a graph showing grain displacement from an equilibrium position for a solitary wave traveling through a column of aligned grains.

Fig. 2b is a graph showing grain velocity for a solitary wave traveling through a column of aligned grains.

Fig. 2c is a graph showing grain acceleration for a solitary wave traveling through a column of aligned grains.

Fig. 3 is a graph showing positions of five topmost grains as a function of time assuming an initial velocity condition v_0 =67.0 m/s produces a solitary wave which manages to extract the surface grain. The dashed region of Fig. 3 represents the liquid and the surface force acts between -50 Angstroms and 0 Angstroms.

Figs. 4a-4c are graphs showing positions of five topmost grains as a function of time. Ten pulses are sent at 0.1 ns apart (corresponding to frequency of 10 GHz), with initial velocity of v_0=37.5 m/s (Fig. 4a), v_0=42.0 m/s (Fig. 4b) and v_0=50.0 m/s (Fig. 4c). One, two or three grains are ejected.

Figs. 5a-5c are graphs showing positions of five topmost grains as a function of time. Ten pulses are generated with initial velocity v_0=50.0 m/s at frequencies of 8GHz (Fig. 5a), 9GHz (Fig. 5b) and 10 GHz (Fig. 5c).

Figs. 6a-6c are graphs showing positions of five topmost grains as function of time. Initial velocity and frequency are kept fixed at v_0=50.0 m/s and ν=10GHz, and the number of pulses is varied at three pulses (Fig. 6a), six pulses (Fig. 6b), and ten pulses (Fig. 6c) to eject the desired number of grains.

Fig. 7 is a graph showing total kinetic energy, total potential energy and total energy of the grain column as a function of time. The initial steps in total energy are due to initial pulsing at the bottom of chain. The final step represents an ejected grain, which returns the "extraction work" to the liquid.

Fig. 8 is a three-dimensional graph illustrating the minimum initial velocity v_0 required to eject "m" grains using "n" pulses. The values for n less than five are not plotted, since they are exceedingly high.

Best Mode(s) for Carrying Out the Invention

Ferrofluids are colloidal systems that are composed of solid, magnetic, single-domain grains in a non-magnetic solvent such as water or oil. The grains are usually coated with a molecular layer, where the molecules are those of the host solvent. Ferrofluid grains are typically between 3 and 15 nm in diameter. Larger ferrofluid particles can be made and can be used for making inks. In a preferred embodiment, this invention relates to dilute ferrofluids in which ferrofluid grains can be regarded as approximately spherical, elastic objects, although other shapes are possible within the spirit of the invention. It is likely that the invention can be generalized for ferrofluids in which the grain shapes possess some irregularities. Instead of ferrofluids, electrorheological fluids, where the grains are responsive to an electric field instead of a magnetic field, may also be used to design an inkjet printer without nozzles.

In a preferred embodiment, we regard grains of 100A diameter as approximately elastic objects in the sense that grain-grain repulsions are Hertzian in nature.

In the description which follows, the term "solitary wave" is defined to be a solitary wave comprising a non-dispersive bundle of mechanical energy, a lump of energy that does not spread out. Solitary waves occur naturally in select systems. Chains of elastic beads are one such system. For more detailed information about solitary waves, first discovered by John Scott Russell in 1834, reference is made to the Heriot-Watt University at Edinburgh web site at www.ma.hw.ac.uk and to R.K. Bullough, "The Wave" "par excellence", the solitary progressive great wave of equilibrium of the fluid - an early history of the solitary wave, in **Solitary waves**, ed. M. Lakshmanan, Springer Series in Nonlinear Dynamics, 1988, 150-281, both of which are incorporated herein by reference.

Certain ferrofluid grains, such as γ-Fe_2O_3 ferrofluid grains which are ~ 8.5 nm in diameter, contain magnetic moments of about $2x10^4\mu_B$. When a ferrofluid containing these grains is subjected to a strong, homogeneous magnetic field B that is directed perpendicular to the ferrofluid surface, the ferrofluid grains tend to align in vertical chains. It has been determined that γ-Fe_2O_3 ferrofluid grains can be suspended in water and one can make a stable ferrofluid. Since water can be colored using dyes, γ-Fe_2O_3 ferrofluid and similar systems can potentially be used as inks.

If one can design a nozzle-free ink-jet printer where the sizes of the ink droplets are dictated by the sizes of the ferrofluid grains rather than by the nozzle diameters that inevitably limit the resolution of ink-jet printers, it may become possible to design inkjet printers of unparalleled resolution using certain ferrofluids. Such printers have the potential to be significantly faster than existing laser printers and may be used for a variety of applications. Such applications may include making very small imprints on devices, making ultra-dense microfiche, to legitimize financial transactions and in marking high denomination currency bills to prevent counterfeiting.

The primary obstacle to designing a ferrofluid printer lies in our inability to extract ferrofluid grains from the surface of a ferrofluid. At first, it might seem that applying a strong static or time varying magnetic field to a dense ferrofluid might be sufficient to extract droplets out of the system. In fact, however, no matter how strong the B field is and what its time dependence may be, it is not possible to extract ferrofluid grains from a ferrofluid. In every case, it turns out that the droplets that one can extract are macroscopic and are inappropriate for the purposes of making "nanodroplets". Thus, it is inconvenient and perhaps undesirable to use a time varying field for extracting ferrofluid grains from a ferrofluid.

We hereby suggest that it may be possible to extract the single ferrofluid grain located nearest to the liquid-air interface by imparting an appropriate impulse at the base of a container in accordance with an embodiment of the invention shown in Fig 1. More specifically, Fig. 1 generally depicts a printer apparatus 10 comprising a container 12 for holding a colloidal liquid 14 having ferrofluid grains 16. Container 12 includes a plurality of nanopores 18 open at the top to provide an exit surface defined by a liquid -air interface. Container 12 preferably includes filler channels 20 for supplying liquid 14 to the container. A magnetic field 22 acts in the direction of the schematic arrow to align the grains 16 into columns extending parallel to the direction of the magnetic field. The magnetic field can be established using known means, for example using an electromagnet or a permanent magnet. A striker plate 24 is located underneath container 12 for providing impulse energy to the bottom of the container. In the present embodiment, striker plate includes an embossed print character 26. In order to free grains 16 residing near the top exit surface of fluid 14, the embossed character 26 of striker plate

24 impacts the bottom of container 12 in a controlled manner to impart energy to the grain columns necessary for overcoming surface forces. A suitable medium 28 is arranged to receive the ejected grains. The liquid 14 is preferably dyed a chosen color, whereby grains 16 appear the color of the dye.

We now show via dynamical studies that it is possible to preferentially eject a desired number of ferrofluid grains that reside near the liquid air interface from any chain in a dilute ferrofluid. The ejection can be effected via trains of non-linear acoustic pulses generated very rapidly at the base of the container. Pulsing may be able to accomplish multiple grain ejection.

Magnetic grains interact with each other via dipolar interactions,

$$V(r_{ij}) = -\Sigma_i\, \mu_i.B + \Sigma_{i>j}(U_{ij}^{dd} + U_{ij}^{nm}), \tag{1}$$

where

$$U_{ij}^{dd} = -(\mu_0^2/r_{ij}^3)[\,\underline{\mu}_i.\underline{\mu}_j - 3(\underline{\mu}_i.\,\underline{r}_{ij})(\underline{\mu}_j.\,\underline{r}_{ij})], \tag{2}$$

and

$$U_{ij}^{nm} = \varepsilon[exp(-[r_{ij} - d]/\eta - exp(-[r_{ij} - d]/2\eta)]. \tag{3}$$

In (2) and (3), $r_{ij} \equiv |r_i - r_j|$. In (1), the first term describes the fact that the magnetic moments $\mu_i = \mu_0\, \underline{\mu}_i$ (where $\underline{\mu}_i$ and $\underline{r}_{ij}$ denote appropriate unit vectors) of the ferrofluid grains tend to align along the field direction to minimize the total energy of the system. The second term, U_{ij}^{dd} describes (see (2)) the interaction between two grains or dipoles in the ferrofluid system. This term can result in repulsion or attraction between the dipoles depending upon the relative orientations of the dipoles. The term U_{ij}^{nm} describes (see (3)) the non-magnetic inetractions between the grains due to a soft-core repulsion at very short distances and a weak short-range attraction. We define $d = 2R$, where R is the radius of each grain in (3). In addition to the forces mentioned above, if and when the grains touch one another, as can be the case when a strong, homogeneous magnetic field is applied to the system, it is likely that there would be a repulsive force that would come into play. If the grains can be regarded as elastic materials, this force would be Hertzian in nature and can be described as follows:

$$V(\delta_{i,i+1}) = (2/5D)(R/2)^{0.5} \equiv a\delta_{i,i+1}^{\;5/2} \tag{4}$$

where $\delta_{i,i+1} = d - (r_i - r_{i+1})$ and $D=(3/2)[\,(1-\sigma^2)/Y]$. The quantities Y and σ refer to the Young's modulus and the Poisson's ratio of the material that makes up the grains.

The typical numbers that are appropriate for studying ferrofluids are as follows. The grain diameter d is taken to be 100 Angstroms or 10^{-8} m, mass is set to be $1.64x10^6$ a.m.u. = $2.72x10^{-21}$ kg and $\mu_0 = 2.1x10^4 \mu_B$ where the Bohr magneton $\mu_B = 9.27x10^{-24}$ J/T. The energy of the dipolar interactions between the neighboring particles separated by $r_{ij} = d$ is given by $\mu_0^2/d^3 \approx 7.50x10^{-21}$ J. Although ferrofluid grains are very large compared to atomic particles, it may be noted that gravity plays a negligible role, being about $2.67x10^{20}$ N on each grain, some eight-orders of magnitude weaker than the dipole-dipole magnetic force, which is about $2.28x10^{-12}$ N.

A typical choice for η in (3) is $2.5x10^{-10}$ m and for ε is $8x10^{-3}$ eV = $1.28x10^{-21}$ J. In (4), we take $Y = 1.0x10^{11}$ Nm$^{-3/2}$ and $\sigma = 0.3$, which are reasonable numbers for the grains, given that the precise values of Y and σ are unknown for most ferrofluid grains. Using the above numbers, we find $a = 1.47x10^6\ Nm^{-3/2}$.

An additional quantity of interest is the surface tension of the water, which is approximately $7.3x10^{-2}\ N/m$. Our model for the surface force that might be experienced by a ferrofluid grain in the vicinity of the surface while trying to escape from the liquid is as follows. In equilibrium, there is no surface force on the surface grain. When the surface grain tends to move above the liquid surface, we assume that the force increases linearly with displacement from the equilibrium position, with the maximum value of the force being $2\pi r\gamma$, which is the maximum static surface force that can be experienced by the macroscopic grain. We assume also that the "extraction work" which is required in order to remove one grain from the fluid is $\pi r^2\gamma$. Since the work done by the surface force should equal the extraction work, we can compute a corresponding "escape distance." If the displacement of the ferrofluid grain exceeds this distance, the surface force becomes zero and the ferrofluid grain is "free". If the surface grain escapes, the surface force is applied to the subsurface ferrofluid grain and then one must determine whether the subsurface grain possesses enough energy to escape and so on.

For ejecting ferrofluid grains through the water-air interface one must overcome a surface force of about $2.29x10^{-9}$ N. For relative grain compression of about 1 Angstrom, the Hertzian forces are of the order of 10^{-8} N and can therefore be an excellent candidate for the purposes of ejecting grains through the liquid-air interface.

The physical structure of a dilute ferrofluid system at strong, vertical (i.e., along the c-axis), homogeneous B fields (say ~ 200 Gauss or $2x10^{-2}$ T or so) can be described as follows. The ferrofluid grains tend to align in chains. The chains can be thought of as vertical stacking of the grains with extremities at the base of the ferrofluid and at the water-air interface (see Fig. 1). It turns out that for dilute ferrofluids, the non-magnetic force, which becomes important at rather high densities of grains in water, is not important and can be ignored. The net effect of the magnetic forces at strong fields is to align the ferrofluid grains in vertical columns along the field. The grains, which are dipoles, can be thought of as touching one another. At equilibrium, the dipole-dipole forces acting on the grains cancel each other, excepting the unbalanced forces acting on the grains at the extremities of the column, which produce a constant loading of the column.

Thermal effects can be ignored at strong magnetic fields. Thus, we assume the columns to be rigid for the purposes of our analyses. This assumption, which is a good first approximation, will be relaxed in more extensive future analyses where the dynamics of the liquid itself will be addressed along with that of impulse propagation in 3D columns.

Nonlinear Dynamics, Solitary Waves and Ejection of the Surface Grain

We briefly mention the problem of ejection of a grain that resides in a column and is nearest to the liquid surface (hereafter referred to as the surface grain). The ejection can be accomplished by sending a non-linear acoustic impulse directed vertically upward at the bottom of the container.

We note that the dipole-dipole attraction between two adjacent ferrofluid grains in each chain is $\propto 1/r^{4}$. Because of the strong dependence of the dipolar force on distance and because the dipolar forces are three orders of magnitude weaker than the Hertzian forces, we consider only the nearest neighbor interactions between the grains in each chain. We have analyzed the dipolar chains using longer-range dipolar forces and have found that such refinements do not significantly affect the calculations to be described below. The net effect of the dipolar forces is to produce a constant magnetic loading of the column, of about $7.3x10^{-3}$ *Angstroms.*

The equation of motion of a spherical ferrofluid grain from the interior of the column, labeled i at location r_i and moving with acceleration d^2r_i/dt^2 can be written as

$$md^2r_i/dt^2 = k\,[\,(d - \Delta_0 - r_i + r_{i-1})^{3/2} - (d - \Delta_0 - r_{i+1} + r_i)^{3/2}\,] + 6\mu_0^2[1/(r_{i+1}-r_i)^4 - 1/(r_i-r_{i-1})^4] \qquad (5)$$

where $k = 5a/2$ and quantity Δ_0 gives the distance of closest approach between the grains in the absence of the pulse and is a parameter that describes the "loading" of the column.

We generate a pulse by imparting an initial velocity v_0 to the first grain at the bottom of the column.

For strong pulses, the second part in (5) (the magnetic interaction) is negligible as compared with the Hertzian contact forces. It was first shown by Nesterenko [8] that such an equation admits solitary wave solution. More recent studies (we cite the publication, S. Sen, M. Manciu and J.D. Wright, "Soliton-like pulses in perturbed and driven Hertzian chains and their possible applications in detecting buried impurities," Physical Review E vol. 57, pp. 2386-2397 (1998)) indicate that a true solitary wave is transmitted through the chain only if $\Delta_0 \to 0$, but even for non-zero loading, the pulse propagation can be very well approximate by the solitary wave, if the compression generated by the pulse largely exceed the initial loading. The formation of solitary waves in chains of elastic beads has also been observed experimentally (we cite A.N. Lazaridi and V.F. Nesterenko, "Observation of a new type of solitary wave in a one-dimensional granular medium," Journal of Applied Mechanics and Technical Physics vol. 26, p. 405 (1985), C. Coste, E. Falcon and S. Fauve, "Solitary waves in a chain of beads under Hertz contact." Physical Review E vol. 56, p. 6104 (1997) and in E.J. Hinch and S. Saint-Jean, "The fragmentation of a line of balls by an impact," Proceedings of the Royal Society of London A Vol. 455, p. 3201 (1999)).

Figs. 2a-2c present plots of the normalized displacement and the corresponding velocity and acceleration functions of the grains versus the position of the grains, in an inertial frame of reference that moves at the speed of the solitary wave. The position of the center of the solitary wave is shown as sitting on grain number *0* to illustrate the symmetry properties of the solitary wave. It is interesting to see that in our study, the kinetic energy of the solitary wave is concentrated within the central grain. The adjacent grains, while a part of the traveling solitary wave, carry a very small fraction of the total

kinetic energy carried by the solitary wave. Therefore, using solitary wave propagation is a natural way to concentrate all the available energy to the topmost grain, which should be ejected from the liquid.

It may be noted that granular contacts typically involve energy loss due to restitution. We define, $w = F_{unloading}/F_{loading} < 1$. Thus, $w < 1$ if restitution is present. We find that the net effect of restitution is to attenuate the amplitude of the traveling impulse rather than to broaden the impulse and make it dispersive (M. Manciu, S. Sen and A.J. Hurd, "The propagation and backscattering of soliton-like pulses in a chain of quartz beads and related problems: (I) Propagation," Physica A (Amsterdam) vol. 274, p. 588 (1999).. In studying the ferrofluid problem, we are interested in the regime in which impulses can generate solitary waves.

Numerical Study of Controlled Grain Ejection

We solve (5) numerically using the Gear algorithm. The initial conditions we use in our calculations are as follows: $dr_1/dt\,|_{t=0} = v_0$ m/s., $dr_i/dt\,|_{t=0} = 0$ for $i>1$ where $i = 1$ defines the grain that is nearest to the striker plate 24 in Fig. 1, i.e., the bottom grain. As mentioned before, the equilibrium positions of the grains are obtained by balancing the dipolar and Hertzian forces and these forces yield a column of grains with constant loading. A perturbation to an edge, which produces compression much bigger than initial loading, results in a solitary wave that propagates from the bottom of the chain to the surface grain at the water-air interface. Solitary waves are strongly non-linear objects and hence their velocities are related to their amplitudes. Our dynamical analyses indicate that solitary wave velocity $c \propto A^{1/4}$, where A is the displacement amplitude of a given solitary wave. For ferrofluid grains of diameter 100 Angstroms, we find that $v_0 \approx 67.0$ m/s or higher for the ejection of the ferrofluid grain which is nearest to the water-air interface. Fig. 3 presents the positions of the first five grains from surface as function of time. The surface force starts to act on grains when their position is 50 Angstroms and has maximum value for $r_i \to 0$ Angstroms. It can be seen that the pulse generated with v_0=67m/s produces the ejection of the topmost grain. It is important to note that in the present approach, an impulse that is some 50 times larger is needed to successfully eject the second particle. Thus, it is energetically highly inefficient to use single impulses to

simultaneous eject multiple grains from the ferrofluid. For the purposes of designing an inkjet printer using ferrofluids, it may be desirable to eject a chosen number of grains from the liquid. As mentioned above, each grain preferably contains a coating of the host liquid. If the water based host liquid is colored, then depositing several grains on some surface might help make a dot with sharper color and hence better visibility.

One possibility would be to send pulses after the columns relaxes from each topmost grain ejection. Since the relaxation time is of the order of 10^{-8}s, a successful pulsing should be in the MHz range. A numerical analysis of this process imply a more complex model, in which the dynamics of liquid itself will be addresses along with that of impulse propagation in a 3D column of grains.

Herein, we explore the possibility of controllably ejecting multiple grains from the ferrofluid before the system relaxes. The physical idea underlying this approach is as follows. One must drive the column in such a way that a set of solitary waves can successively reach the end of the column, which is near the liquid-air interface. It is envisioned that the combined effect of the clustered solitary waves could lead to the near simultaneous ejection of multiple grains from near the surface. As we shall see, it turns out that such pulsing indeed leads to multiple grain ejection in our model based analyses.

In our calculations we consider three key parameters: (i) the initial velocity of the first grain due to the pulse, v_0, (ii) the number of pulses generated, and (iii) the frequency that characterizes how often the impulses are being initiated at the bottom grain of the chain of ferrofluid grains, ν. Since the grain velocity is of the order of 10m/s, the time taken for passing the liquid-air interface for a grain is about 10^{-9}s, which implies that in order to have multiple pulses interfering at surface, one should pulse the system with frequencies of the order 10 GHz.

Figs. 4a-4c present the displacement of the first five grains from surface. A train of ten pulses was sent through the chain by resetting the velocity of the bottom grain to v_0 at fixed intervals of time (0.1 ns). For v_0=37.5m/s (Fig.4a) only the surface grain is ejected; by increasing v_0 to 42.0 m/s and 50.0 m/s, two (Fig.4b) or respectively three grains (Fig.4c) can be ejected.

A similar study is shown in Figs. 5a-5c, where ten pulses are generated using constant impact velocity v_0 =50.0 m/s. By changing the frequency of pulsing, one or

more grains can be simultaneously ejected. The frequencies used are 8 GHz for one particle ejection (Fig.5a), 9GHz for two particle ejection (Fig.5b) and 10 GHz for three particle ejection.

Since the fine-tuning of the pulsing frequency or controlling the energy of each pulse might be quite challenging in practice, more important for application purposes is the dependence of number of ejected grains to the number of pulses. We repeat the study for v_0=50.0 m/s and frequency of 10 GHz (time window of 0.1 ns between pulses). As Figs. 6a-6c show, it takes three, six, or ten pulses in order to eject one, two or three particles, respectively.

An important issue is the numerical accuracy of integration procedure. Fig. 7 presents the kinetic, potential and total energy as function of time for a typical calculation in which ten pulses was generated at the bottom of the column and one grain is ejected from the liquid. The final step of total energy represents the "extraction work" for the grain (energy restituted to the liquid after ejection, which therefore disappears from the chain energy). While total energy is constant with accuracy better than 0.01%, the kinetic and potential energies show chaotic behavior after the first solitary wave reaches the top of the chain. This indicates that, although it is straightforward to integrate numerically the equation of motion, it is very unlikely to obtain an analytic form for the motion of the topmost grains.

It should be mentioned that the results in Figs 4-6 are just sample calculations and not optimal values. For the model system described in this application, one can ask the question what is the minimum pulsing velocity, such as to eject simultaneous "m" grains using "n" pulses. This question could be answered by careful integration of the equation of motion for many initial conditions; our results are summarized in Fig. 8. The structure of the "staircases" shown is complex and more studies are needed to understand its geometry as v_0 and number of pulses are varied.

Figures 4-8 provide insights into the fact that it is possible to eject the grain nearest to the liquid-air interface using much smaller hit velocities than that in the single impulse case (where $v_0 = 67.0$ m/s). To accomplish this, one needs to use a series of high frequency impulses with tailored amplitudes.

Our computation shows that a desired number of grains can be ejected if the initial hit speed v_0 possesses the appropriate magnitude. This magnitude is typically between a few m/s and some *60* m/s for grains with diameter of 100 Angstroms.

Scaling of v_0 with Grain Size

Let us consider spherical grains of some standard radius r_0 and let s be a scale factor to control grain radius $r = sr_0$. Then $m = s^3m_0$, $F_m = s^2F_{m0}$, $F_g = s^3F_{g0}$ and $F_s = sF_{s0}$. The last scaling relation implies that $D_{esc} = sD_{esc0}$. In the absence of restitution, the initial kinetic energy imparted to a column, $E_0 = mv^2/2$ must equal the minimum energy required for escape, $E_{esc} = \pi r^2\gamma = s^3m_0v^2/2 = \pi s^2r_0^2\gamma = s^2E_{esc0} = s^2m_0v_0^2$. Thus, $v^2 = v_0^2/s$ or $v = s^{-1/2}\ v_0$. The particle dynamics based results in Figure 5 confirm the validity of this scaling law, yielding an exponent of 0.497.

Comments on Polydispersity in Chains

One may worry that even the best available ferrofluid grains are seldom monodisperse and whether the predictions made herein remain valid in the presence of polydispersity. It was shown by Spence in Proceedings of Royal Soceity of London A vol. 305, p. 55 (1968), that the index of the Hertz law depends upon the geometry of the grain-grain contact. Typically, this index lies in a range that runs roughly between 3/2 and 2. We have studied the propagation of solitary waves in chains of same r but with small (few percent) random variation in the index around 3/2 and 2. Our results reveal that the width of the solitary wave is weakly dependent upon the index in the above-mentioned range of values and hence the solitary wave is at best marginally affected by variations in grain contacts. The solitary wave is more significantly affected when the radius r varies significantly. Thus, approximately spherical ferrofluid grains, which possess roughly the same radius, should be appropriate for our purposes.

<u>Industrial Applicability</u>

We studied a simplified model for the magnetically ordered dilute ferrofluid and we shown that it may be possible to carry out controlled ejection of ferrofluid grains from a ferrofluid using nonlinear pulsing at the bottom of the container.

Ferrofluid grains carry a coating of a thin layer of the host liquid. Water based host liquids can be easily colored. Hence, ferrofluid grains can be useful to produce "nanodrops" of inks. Such drops would be at least an order of magnitude smaller than the state-of-the-art in inkjet printing.

The ejected grains can be forced to impinge on an object of choice such as a piece of paper or a surface. Using appropriate designing, one can use the ink-coated ferrofluid grains to print letters and symbols. Further, since the velocity of the ferrofluid drop can also be controlled, it may be possible to use these drops to print on a variety of surfaces.

Moreover, using appropriate designing, one can use the ink-coated ferrofluid grains to print letters and symbols.

Thus, it is seen that the objects of the invention are efficiently obtained. The description of the preferred embodiment should not be regarded as limiting, as modifications and changes in the invention should be readily apparent to those having ordinary skill in the art, and these modifications are intended to be within the spirit and scope of the invention as claimed.

Claims

1. A method for ejection of dipolar grains from a colloidal liquid comprising the steps of:

holding said liquid in a container to provide said liquid with an open exit surface from which said grains can be ejected;

applying a homogeneous field to said liquid to align said grains in a plurality of columns extending substantially normal to said exit surface of said liquid; and

initiating a non-linear acoustic impulse in said liquid, said impulse propagating in a longitudinal direction of said plurality of columns;

whereby at least one grain from at least one of said plurality of columns is forced out of said liquid by said impulse.

2. The method for ejection of dipolar grains according to claim 1, wherein said liquid is a ferrofluid.

3. The method for ejection of dipolar grains according to claim 1, wherein said liquid is an electrorheological fluid.

4. The method for ejection of dipolar grains according to claim 1, wherein said exit surface is defined by an interface between a gas and said liquid.

5. The method for ejection of dipolar grains according to claim 1, wherein said exit surface is defined by an interface between a vacuum and said liquid.

6. The method for ejection of dipolar grains according to claim 2, wherein said homogeneous field is a magnetic field.

7. The method for ejection of dipolar grains according to claim 3, wherein said homogeneous field is an electric field.

8. The method for ejection of dipolar grains according to claim 1, wherein said acoustic impulse is a solitary wave.

9. The method for ejection of dipolar grains according to claim 1, wherein a plurality of acoustic impulses is initiated to eject a plurality of said grains from said liquid.

10. The method for ejection of dipolar grains according to claim 9, wherein acoustic impulses of said plurality of acoustic impulses constructively interfere with each other near said exit surface.

11. The method for ejection of dipolar grains according to claim 1, further comprising the step of dying said liquid to apply a chosen color to said grains.

12. An apparatus for ejection of dipolar grains from a colloidal liquid comprising:

a container for holding said liquid to provide said liquid with an open exit surface from which said grains can be ejected;

means for applying a homogeneous field to said liquid to align said grains in a plurality of columns extending substantially normal to said exit surface of said liquid; and

means for initiating a non-linear acoustic impulse in said liquid, said impulse propagating in a longitudinal direction of said plurality of columns;

whereby at least one grain from at least one of said plurality of columns is forced out of said liquid by said impulse for receipt by a medium.

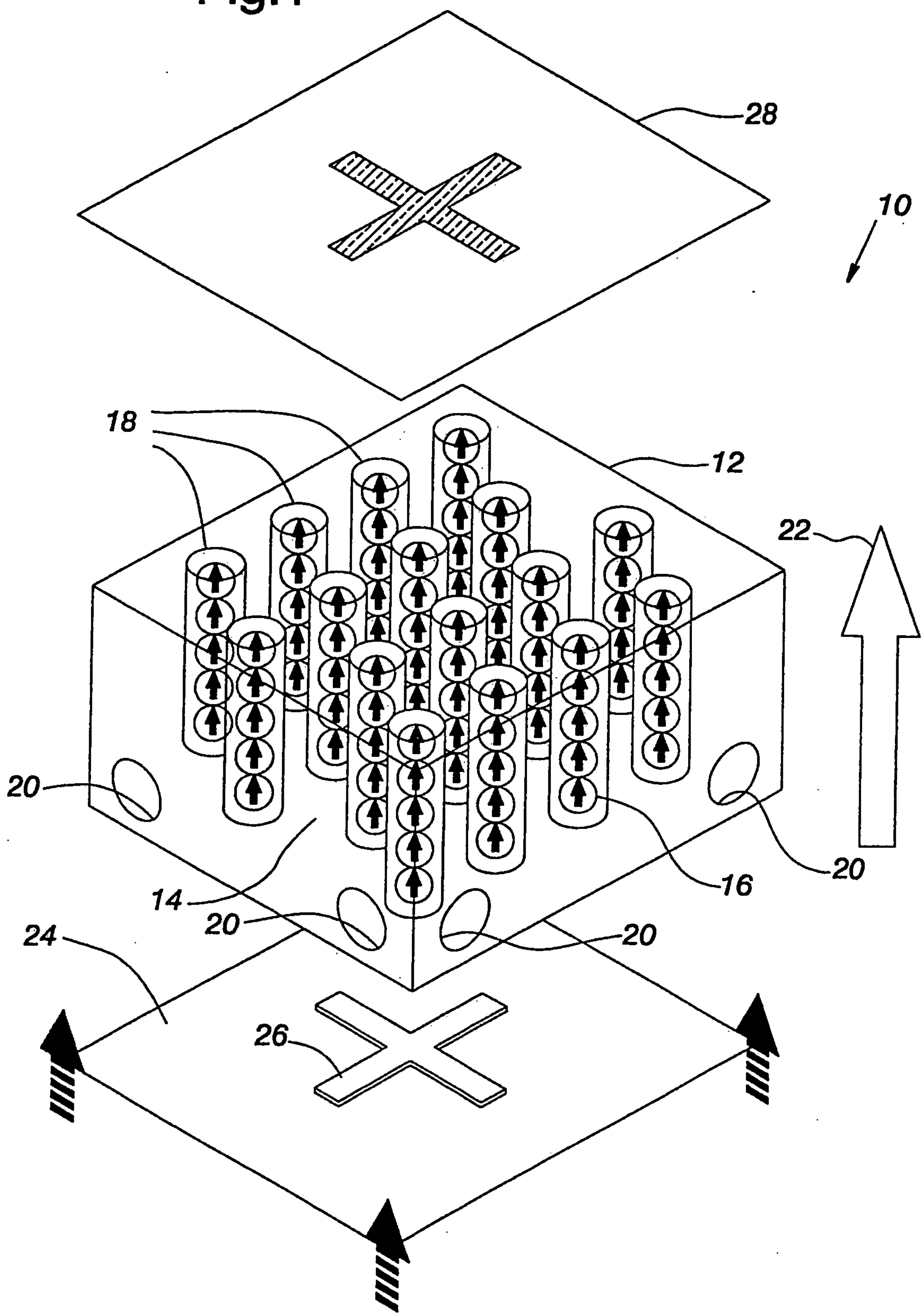
13. The apparatus for ejection of dipolar grains according to claim 12, wherein said colloidal liquid is a dilute ferrofluid.

14. The apparatus for ejection of dipolar grains according to claim 13, wherein said means for applying said homogeneous field is an electromagnet for applying a magnetic field.

15. The apparatus for ejection of dipolar grains according to claim 12, wherein said means for initiating a non-linear acoustic impulse is a striker plate for hitting a bottom of said container.

16. The apparatus for ejection of dipolar grains according to claim 12, wherein said liquid is dyed a chosen color.

Fig.1



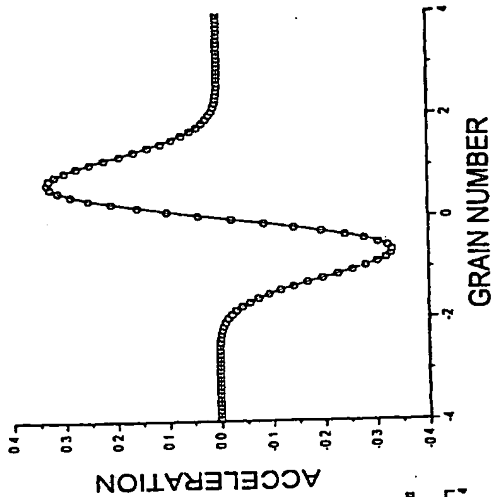


Fig. 2c

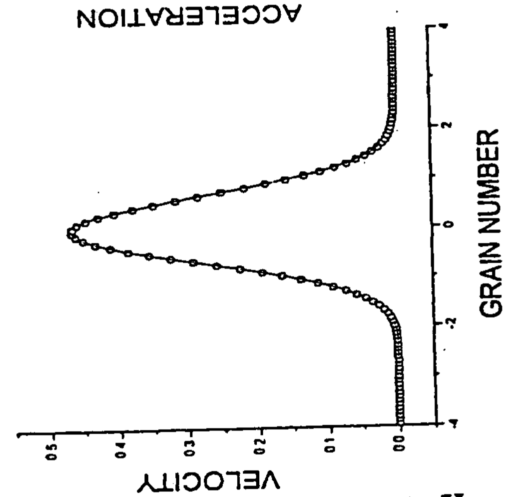


Fig. 2b

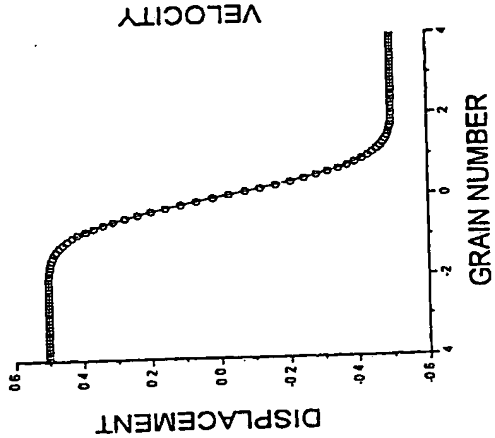
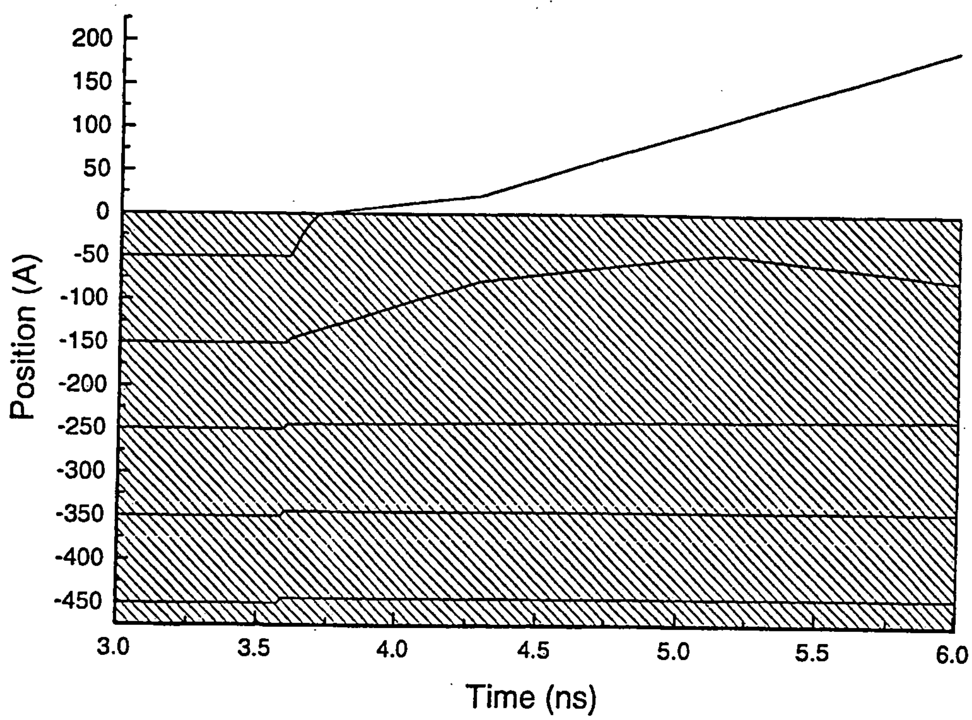


Fig. 2a



200
150
100
50
0
-50
-100
-150
-200
-250
-300
-350
-400
-450
Position (A)
3.0
3.5
4.0
4.5
5.0
5.5
6.0
Time (ns)

Fig. 3

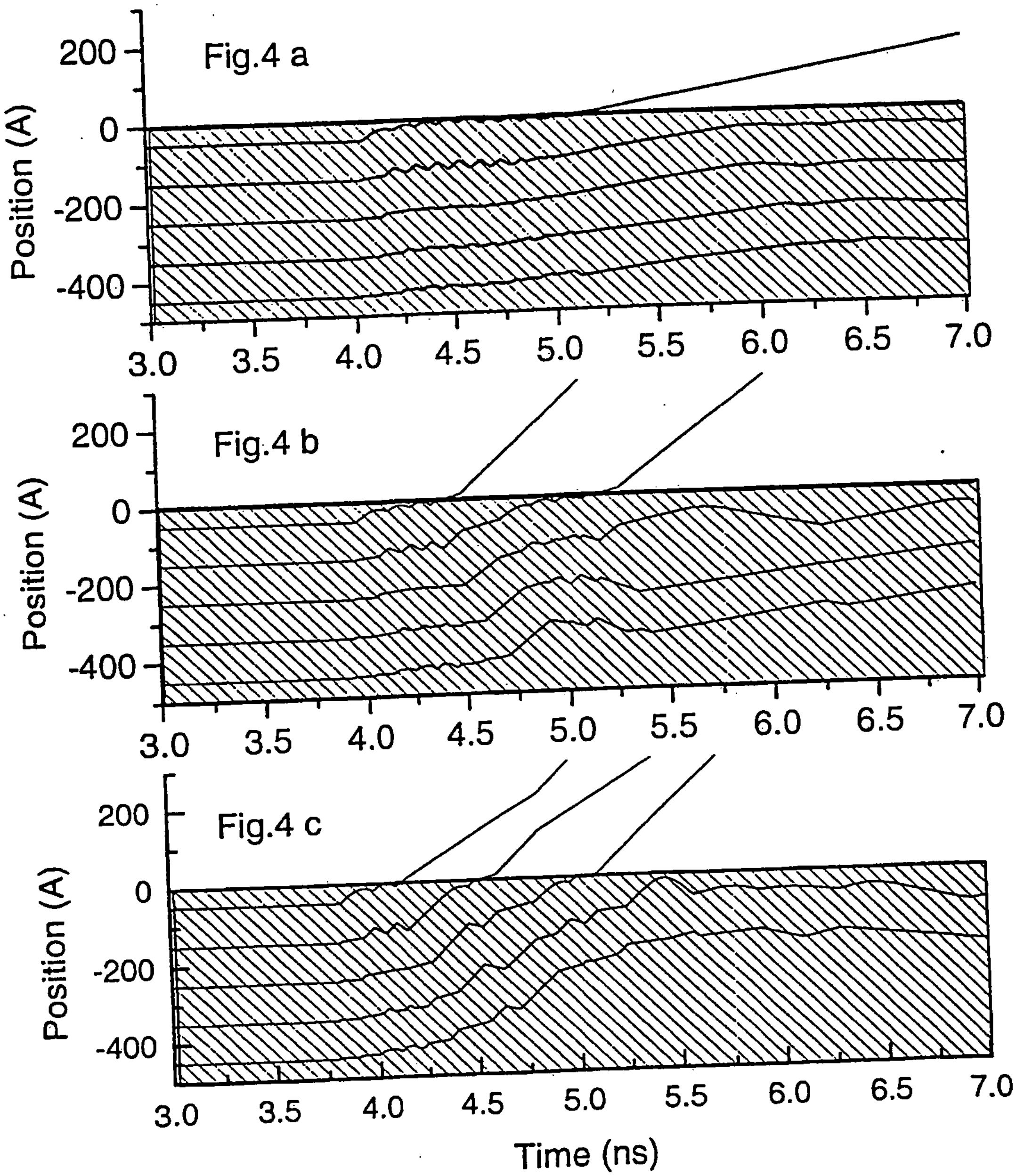


Fig.4 a
Position (A)
200
0
-200
-400
3.0
3.5
4.0
4.5
5.0
5.5
6.0
6.5
7.0
Fig.4 b
Fig.4 c
Time (ns)

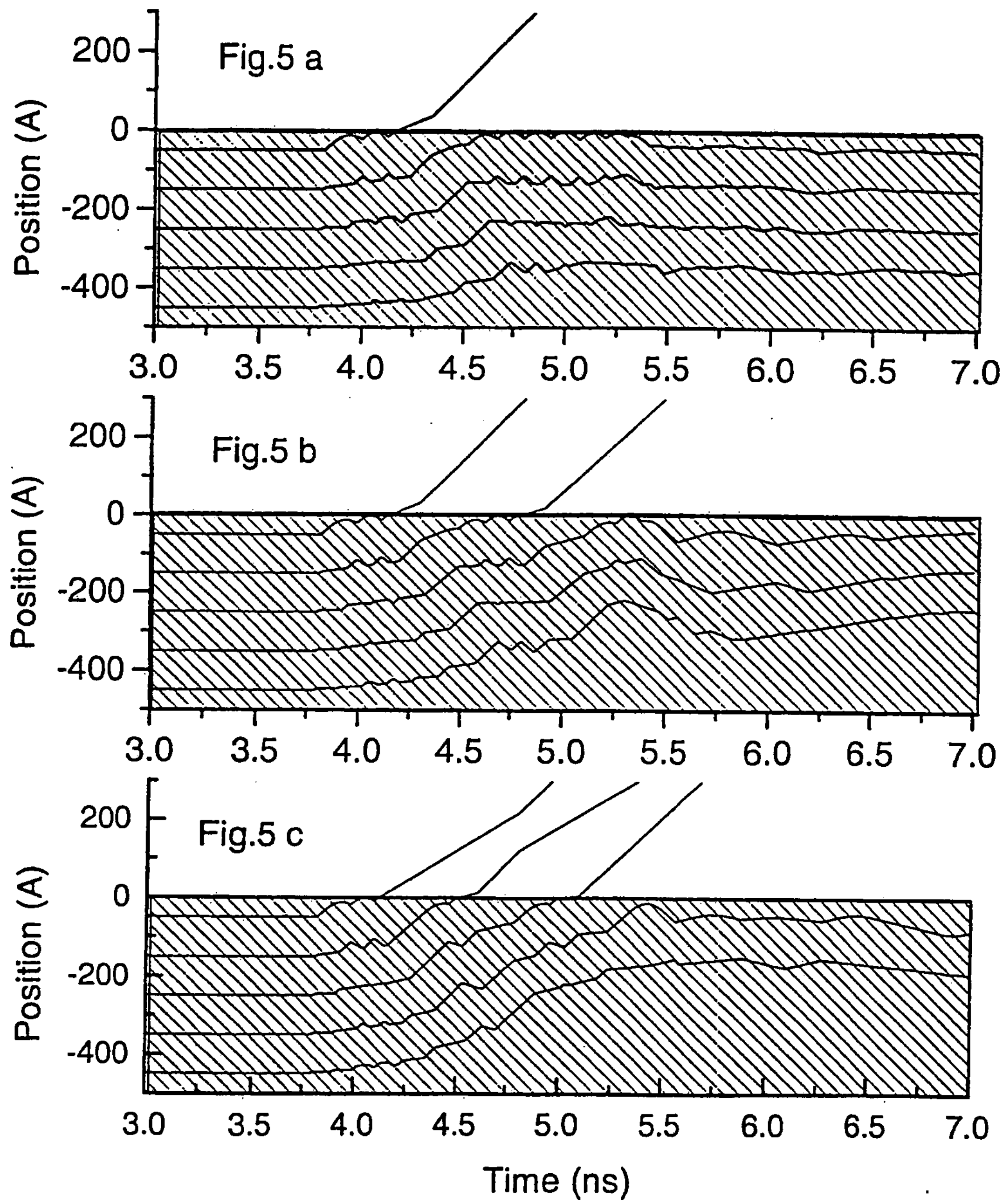


Fig.5 a
Position (A)
200
0
-200
-400
3.0 3.5 4.0 4.5 5.0 5.5 6.0 6.5 7.0
Fig.5 b
Position (A)
200
0
-200
-400
3.0 3.5 4.0 4.5 5.0 5.5 6.0 6.5 7.0
Fig.5 c
Position (A)
200
0
-200
-400
3.0 3.5 4.0 4.5 5.0 5.5 6.0 6.5 7.0
Time (ns)

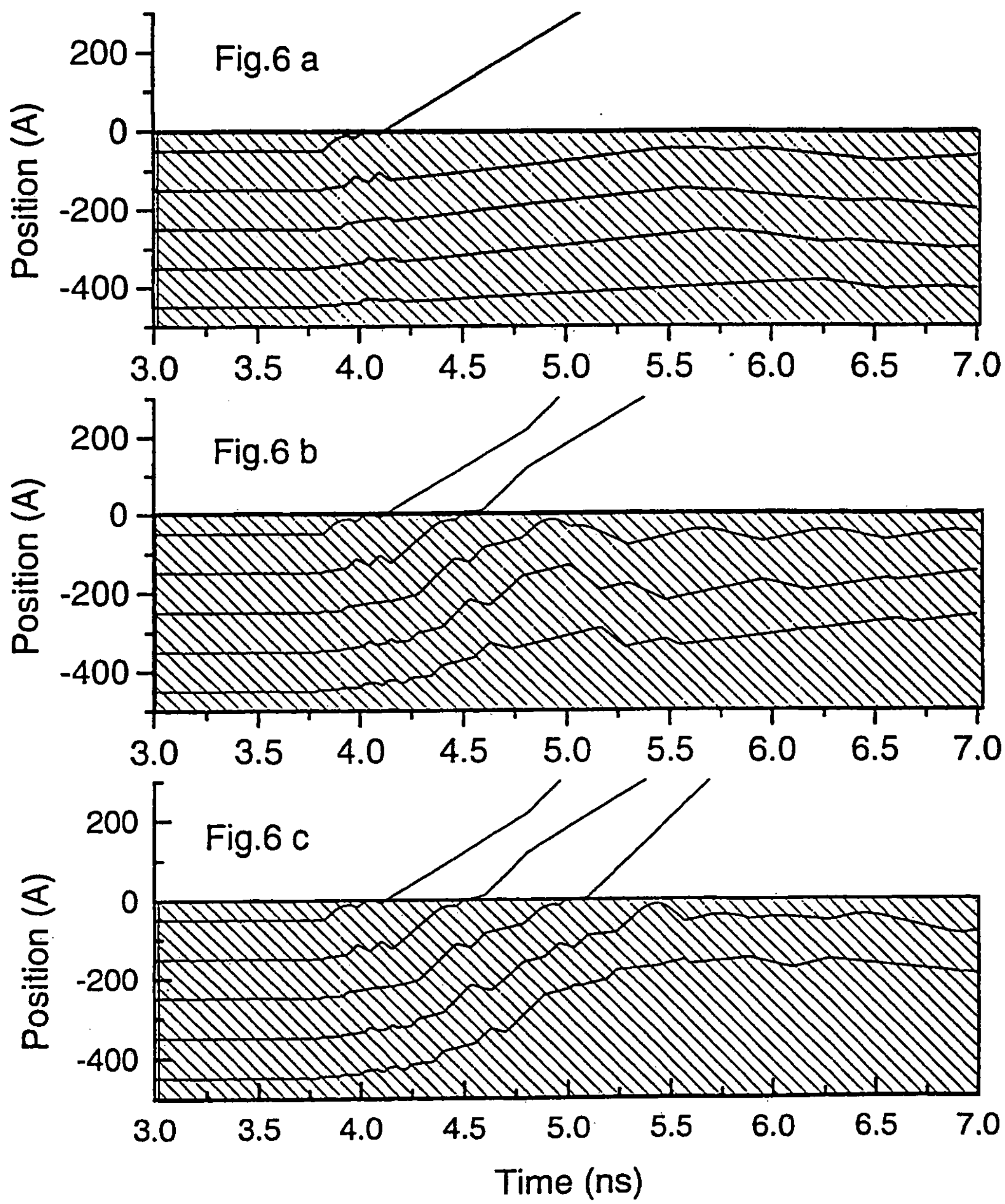
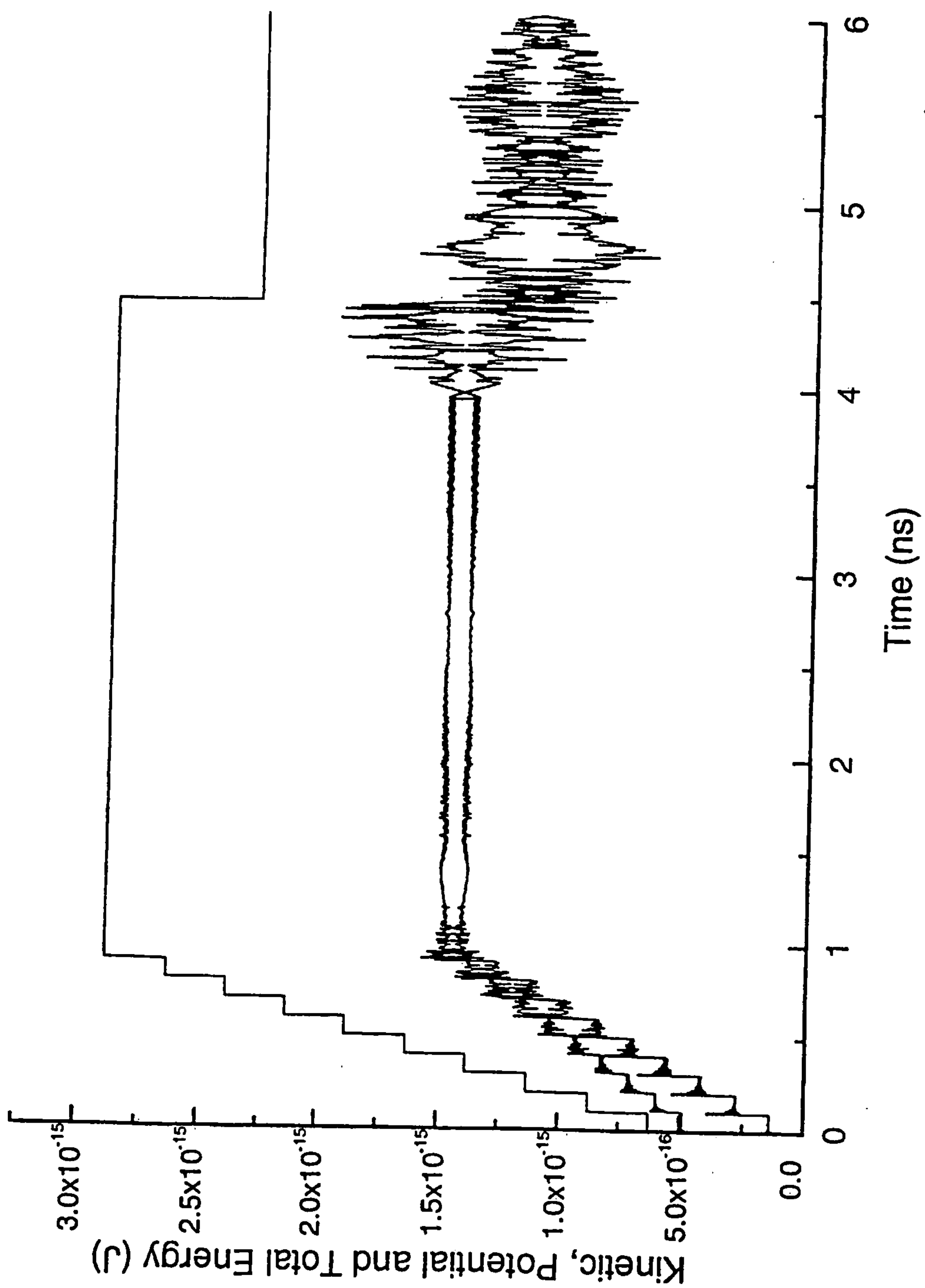
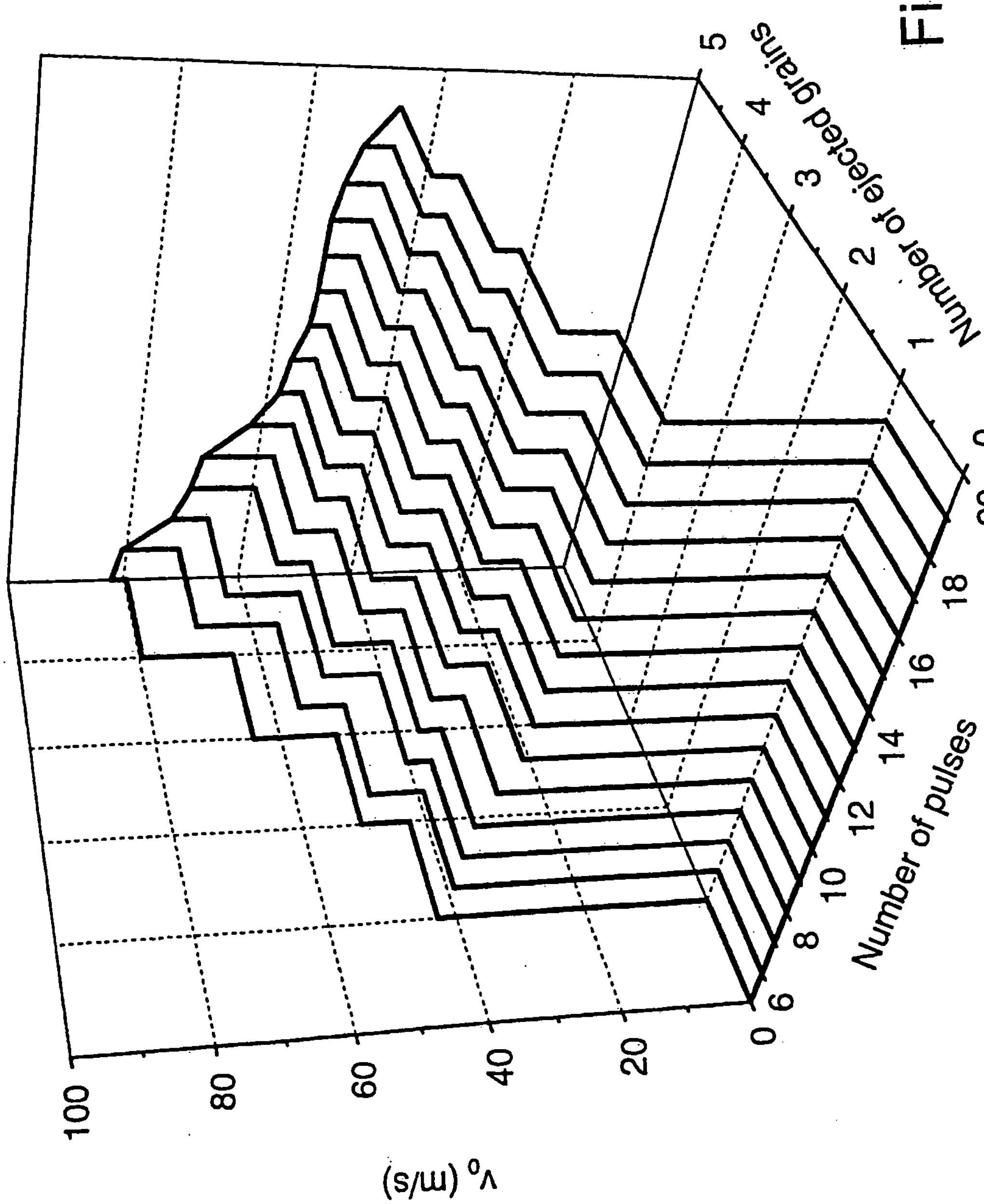


Fig.6 a
Position (A)
200
0
-200
-400
3.0
3.5
4.0
4.5
5.0
5.5
6.0
6.5
7.0
Fig.6 b
Fig.6 c
Time (ns)

Fig. 7


Kinetic, Potential and Total Energy (J)
3.0x10^-15
2.5x10^-15
2.0x10^-15
1.5x10^-15
1.0x10^-15
5.0x10^-16
0.0
Time (ns)
0
1
2
3
4
5
6



Number of ejected grains
0
1
2
3
4
5
Number of pulses
6
8
10
12
14
16
18
20
v_o (m/s)
0
20
40
60
80
100

Fig. 8

INTERNATIONAL SEARCH REPORT

Intern. .al Application No
PCT/US 00/23990

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 B41J2/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 B41J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

PAJ, EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP 07 156400 A (XEROX CORP.) 20 June 1995 (1995-06-20) abstract & US 6 048 050 A (GUNDLACH R.W. AND RAWSON E.G.) 11 April 2000 (2000-04-11) the whole document ---	1,3-5, 7-12,15, 16
A	PATENT ABSTRACTS OF JAPAN vol. 1996, no. 02, 29 February 1996 (1996-02-29) & JP 07 276622 A (CANON INC), 24 October 1995 (1995-10-24) abstract --- -/--	1-16

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
5 December 2000	14/12/2000

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Didenot, B

1

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category °	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 760 286 A (HEWLETT PACKARD CO) 5 March 1997 (1997-03-05) abstract column 7, line 56 -column 9, line 4 figures -----	1,2,4-6, 12-14

1

INTERNATIONAL SEARCH REPORT

Information on patent family members

Intern. nal Application No
PCT/US 00/23990

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
JP 7156400 A	20-06-1995	US 6048050 A	11-04-2000
JP 07276622 A	24-10-1995	NONE	
EP 0760286 A	05-03-1997	JP 9118020 A	06-05-1997